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# Impact of Gouy-Chapman-Stern model on conventional ISFET sensitivity and stability

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## Abstract

Utilizing Gouy-Chapman-Stern model can improve ISFET sensitivity and stability using Stern layer in direct contact with electrolyte in ISFET sensing window. However, this model remains a challenge in mathematical way, unless it's re-applied using accurate simulation approaches. Here, we developed an approach using a commercial Silvaco TCAD to re-apply Gouy-Chapman-Stern model as ISFET sensing membrane to investigate its impact on sensitivity and stability of conventional ISFET. SiO<sub>2</sub> material and high-k Ta<sub>2</sub>O<sub>5</sub> material have been examined based on Gouy-Chapman and Gouy-Chapman-Stern models. Results shows that the ISFET sensitivity of SiO<sub>2</sub> sensing membrane is improved from ~38 mV/pH to ~51 mV/pH and the V<sub>TH</sub> shift stability is also improved. Additionally, the results indicate that the sensitivity of Ta<sub>2</sub>O<sub>5</sub> is 59.03 mV/pH that hit the Nernst Limit 59.3 mV/pH and achieves good agreements with mathematical model and previous experimental results. In conclusion, this investigation introduces a real validation of previous mathematical models using commercial TCAD approach rather than expensive fabrication that paves the way for further analysis and optimization.

**Keywords:** Gouy-Chapman-Stern, high-k, ISFET sensitivity, stability, stern layer

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## 1. Introduction

CMOS technology now provides an opportunity for chemical sensing platforms to leverage semiconductor technology that may offer advantages such as scalability, miniaturisation, fabrication, and integration with intelligent instrumentation. ISFET sensors are the most promising, and may satisfy all these opportunities. Due to its promising application in biological, biochemical and medical detection [1-5], ISFET has received much interest since it was first reported by Bergveld in 1972 [6]. Particularly, much effort has been made to investigate pH sensitive ISFETs with studies on device structures and pH-sensing membranes aimed at improving the sensitivity and stability of ISFETs [7, 8]. It is well known that the gate dielectric is in direct contact with the electrolyte solution, which determines the starting sensitivity of these devices. As the SiO<sub>2</sub> gate dielectric shows a low response sensitivity and poor stability, other inorganic materials such as Al<sub>2</sub>O<sub>3</sub> [8, 9], Si<sub>3</sub>N<sub>4</sub> [7, 10], Ta<sub>2</sub>O<sub>5</sub> [9, 11], HfO<sub>2</sub> [12-15] and ZrO<sub>2</sub> [12-14] with their enhanced stability and sensitivity have also been investigated.

Until now, different models and adaptations of models have been used to model the pH-change sensitivity in ISFETs, the development of surface potential in the area, and in the further course of a Helmholtz double layer (DL). Louis Georges Gouy in 1910, and David Leonard Chapman in 1913, together noticed that capacitance was not a constant, but depended on the applied potential and the ionic concentration. The Gouy-Chapman model made an important contribution by proposing a diffuse model of the DL. In this model, "the charge distribution of ions as a function of distance from the metal surface allows Maxwell-Boltzmann statistics to be applied". Thus, the electric potential decreases exponentially at distances further away from the surface of the fluid bulk. However, the Gouy-Chapman model fails for highly charged DLs. In 1924, Otto Stern suggested combining the Helmholtz model with the Gouy-Chapman model: In Stern's model, some ions adhere to the electrode as suggested by Helmholtz, giving an internal Stern layer, while some form a Gouy-Chapman diffuse layer.

Figure 1 demonstrates schematic representations of electrical double layer structures describing (a) the Helmholtz model, (b) the Gouy-Chapman model and (c) the Gouy-Chapman-Stern model.

This paper extends the previous investigations [16] that aimed at developing a different Models for simulation of ISFET operations. Particularly, in this work we examine more closely the links between IC design simulation and previous mathematical models have not be clearly linked and for more accurate analysis for further optimization.

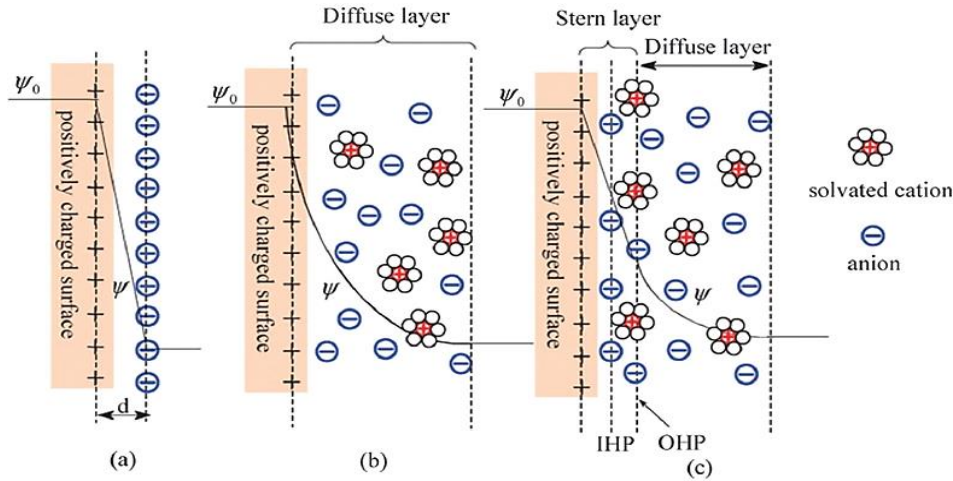


Figure 1. ISFET DL's models [15]

## 2. Material and Methods

### 2.1. Mathematical Model

In the original structure of an ISFET, the gate oxide is in direct contact with the electrolyte solution as shown in Figure 2 (a), acting as a sensing dielectric. The Stern layer is a modified version of the ISFET in which the sensing layer is separated from the gate oxide by using an extended conductive layer Figure 2 (b) after the gate oxide is covered by the electrolyte solution, creating a more robust structure for extended efficacy in the solutions. Through the effective coupling capacitance between the sensor surface and Floating Gate (FG), the surface potential ( $\psi_0$ ) modulates the potential of the FG, and therefore there will be a corresponding shift in the threshold voltage ( $V_T$ ) of the sensor [17-21]. Therefore, from the Site-binding model, the charge density can be expressed by [19, 20, 22]:

$$\sigma_0 = qN_{sil} \left( \frac{aH_s^{+2} - K_a K_b}{K_a K_b + K_b aH_s^{+} + aH_s^{+2}} \right) \quad (1)$$

where;  $aH_s^{+}$  is the  $H^{+}$  activity calculated by ( $aH_s^{+} = 10^{-pH_s}$ );  $q$  is the elementary charge;  $N_{sil}$  is the density of the available sites and  $K_a$ ,  $K_b$  represents the intrinsic dissociation constants, and the  $N_{sil}$ ,  $K_a$  and  $K_b$ , are oxide layer dependent.

Based on "charge density", the charge on the electrolyte side of a double layer ( $\sigma_{DL}$ ) is the same value, but is a negative charge. Therefore, this charge can be calculated from the integral double layer capacitance ( $C_i$ ) and the surface potential [23, 24]:

$$\sigma_{DL} = -C_i \psi_0 = -\sigma_0 \quad (2)$$

Therefore, by solving (1) and (2), we can demonstrate the relation between  $aH_s^{+}$  and  $\psi_0$  parameters. According to the Boltzmann distribution model for the  $H^{+}$  ions [9, 24], the pH value at the sensor surface is [9]:

$$pH_s = pH_B + \frac{q \psi_0}{2.3kT} \quad (3)$$

T and k represent the absolute temperature and Boltzmann constant, respectively. The subscript S and B denote the pH at the sensor surface and in the bulk solution, respectively, and  $\psi_0$  is the potential drop across the diffusion layer. In (3), the intrinsic buffer capacity ( $\beta_{int}$ ) is defined as the ability to collect a charge at the sensor surface ( $\sigma_0$ ) due to the change in surface pH (pHs) [9]:

$$\beta_{int} = \frac{\Delta\sigma_0}{-q \Delta pH_S} \quad (4)$$

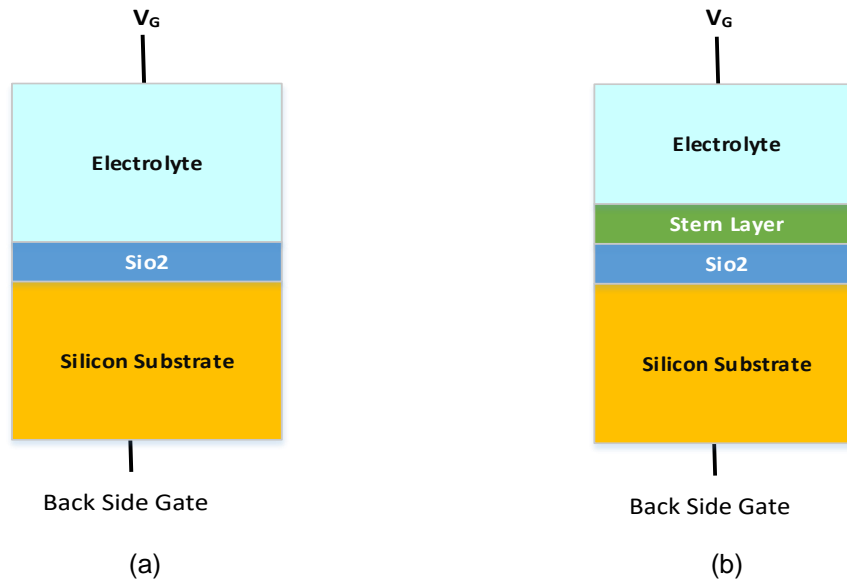


Figure 2. ISFET structure based on (a) Chapman model (b) Gouy-Chapman-Stern model

The diffusion capacity ( $C_{diff}$ ) is the ability to store the opposing charge in the solution near the surface, due to the change in surface potential [9]:

$$C_{diff} = \frac{\Delta\sigma_0}{\Delta\psi_0} \quad (5)$$

therefore, we can write:

$$\frac{\Delta\psi_0}{\Delta pH_S} = -q \frac{\beta_{int}}{C_{diff}} \quad (6)$$

Differentiating (3) with respect to the bulk pH, and using (6), the surface potential sensitivity to the pH<sub>B</sub> can be derived as [9]

$$\frac{\Delta\psi_0}{\Delta pH_B} = -2.3 \frac{kT}{q} \left( \frac{1}{1+\alpha} \right) \quad (7)$$

with

$$\alpha = \frac{2.3 k T C_{diff}}{q^2 \beta_{int}} \quad (8)$$

here,  $\alpha$  is a positive sensitivity (dimensionless parameter). Therefore, the sensitivity of the potential at the sensor surface and the corresponding change of the sensor threshold voltage to the bulk pH are limited to 59.3 mV/pH (Nernst limit). In the Stern model, the relationship between the diffusion layer potential  $\psi_0$  (previous calculations) and the Stern potential  $\psi_s$  can be expressed by:

$$(\psi_0 - \psi_s) = \frac{(8kT\epsilon\epsilon_0 n^0)^{\frac{1}{2}} \sinh\left(\frac{zq\psi_s}{2kT}\right)}{C_s} \quad (9)$$

where  $\epsilon\epsilon_0$  is the permittivity of free space and its relative permittivity, respectively;  $n^0$  is the number concentration of each ion in the bulk,  $z$  is the magnitude of the charge on the ions and  $C_s$  represents the integral capacitance of the Stern layer.

## 2.2. TCAD Simulation

Commercial TCAD allows users to introduce bias-dependent surface charges in the form of interface donor or acceptor traps. The challenge is simulating the updated surface charge density equation described by in the electrolyte pH change model [19]. To introduce this equation to the simulator, interface trap statements are utilized to mimic the surface charge accurately, as follows [21]:

$$INTTRAP <type> E. LEVEL= <r> DENSITY= <r> <capture parameters>$$

“INTTRAP” activates interface defect traps at discrete energy levels within the bandgap of the semiconductor and sets their parameter values. Device physics has established the existence of three different mechanisms, which add to the space charge term in Poisson’s equation in addition to the ionized donor and acceptor impurities” [21]. Interface traps will add space charge directly into the right-hand side of Poisson’s equation. To calculate the trapped charge in Poisson’s equation, the total charge value is defined by the following:

$$\sigma_0 = q(N_{tD}^+ - N_{tA}^-) \quad (10)$$

where  $N_{tD}^+$  and  $N_{tA}^-$  are the densities of ionized donor-like and acceptor-like traps, respectively. DENSITY and its probability of ionization are represented as  $FtA$  and  $FtD$ , respectively. For donor-like and acceptor-like traps, the ionized densities are calculated by the following equations:

$$N_{tD}^+ = DENSITY \times FtD \quad (11)$$

$$N_{tA}^- = DENSITY \times FtA \quad (12)$$

where  $FtA$  and  $FtD$  are given by the following equations:

$$FtA = \frac{V_n SIGN n + e_{pA}}{V_n SIGN n + V_p SIGP p + e_{nA} + e_{pA}} \quad (13)$$

$$FtD = \frac{V_p SIGP p + e_{nD}}{V_n SIGN n + V_p SIGP p + e_{nD} + e_{pD}} \quad (14)$$

where  $SIGN$  is the carrier capture cross sections for electrons and  $SIGP$  holes. The thermal velocities for electrons and holes are  $V_n$  and  $V_p$ , respectively. For donor-like traps, the electron and hole emission rates,  $e_{nD}$  and  $e_{pD}$ , are defined by the following [21]:

$$e_{nD} = \frac{1}{DEGEN.FAC} V_n SIGN n_i e^{\frac{E_t - E_i}{kT}} \quad (15)$$

$$e_{pD} = DEGEN.FAC V_p SIGP n_i e^{\frac{E_i - E_t}{kT}} \quad (16)$$

where  $E_t$  and  $E_i$  are the trap energy level and the intrinsic Fermi level position, respectively. DEGEN.FAC is the degeneracy factor of the trap center. For acceptor traps, the electron and hole emission rates,  $e_{nA}$  and  $e_{pA}$ , are defined by the following [21]:

$$e_{nA} = DEGEN.FAC V_n SIGN n_i e^{\frac{E_t - E_i}{kT}} \quad (17)$$

$$e_{pA} = \frac{1}{DEGEN.FAC} V_p SIGP n_i e^{\frac{E_t - E_f}{kT}} \quad (18)$$

For example, the acceptor interface trap statement and its parameters are as following:

**INTTRAP ACCEPTOR E.LEVEL: DENSITY: INTMATERIAL: DEGEN.FAC EoN: EoP:**

considering all equations mentioned above, we can rewrite the sit-binding model (1) based on TCAD model. We first assume that acceptor and donor traps exchange carriers only with the conduction and valence band of the semiconductor representing the electrolyte, respectively. Hence, we can rewrite (7) in terms of TCAD model as follows:

$$\sigma_{(TCAD)} = q \times DENSITY \left( \frac{V_p SIGP P - V_n SIGN n}{v_p SIGP P + V_n SIGN n + K_b n_i^2} \right) \quad (19)$$

for more details about TCAD simulation and modelling, the previous work was well described all modelling methodologies [25].

### 3. Result and Discussion

Models and simulation results have been shown in this section. In modeling part, Figure 3 examine the difference between Gouy-Chapman and Gouy-Chapman-Stern models using (8). The sensitivity parameter ( $\alpha$ ) (Dimensionless parameter) have been shown the sensitivity of surface potential changing based on range of pH bulk [21]. The value of  $\alpha$  varies between 0 and 1 depending on the intrinsic buffer capacity and the differential capacitance [8]. Here we converted the values as proportions values from 0 to 59.3 that is the value of top Nernst level. For a sensitivity close to the theoretical maximum,  $\alpha$  approaches 59.3, the intrinsic buffer capacity should be high, and the differential capacitance should be small, as can be concluded from (8). A sensitivity close to zero can theoretically be derived when the intrinsic buffer capacity approaches zero. This shows that the Gouy-Chapman model remain challenge the Roll-off problem that already compensated by Gouy-Chapman-Stern models as we shown in Figure 3.

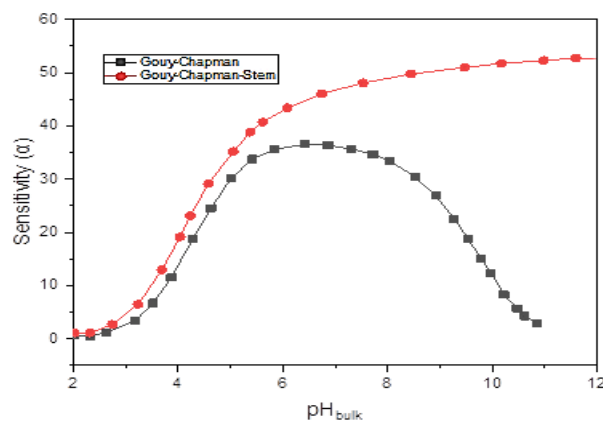


Figure 3. Theoretical sensitivity parameter

The theoretical surface potential shift based on range of pH bulk from pH2-pH12 were observed in Figure 4. The (7) and (9) are the expression for the sensitivity of the electrostatic potential to changes in the bulk pH [9]. As we shown the SiO<sub>2</sub> sensing membrane as Stern layer (9) achieve good agreement with experimental result in respect of sensitivity and stability. Nevertheless, A SiO<sub>2</sub> Gouy-Chapman model still have lack of curve stability as well as low sensitivity. On the other hand, Ta<sub>2</sub>O<sub>5</sub> sensing membrane is behaved well in term of sensitivity

and stability comparing with normal silicon dioxide Which is known because high dielectric constant of high-k material. However, from mathematical models is very clear that using of Gouy-Chapman-Stern model enhance the ISFET sensitivity as well as achieve stable shift in surface potential through pH bulk change. On the other side, TCAD Simulaation results process is contribute same as modeling results. Table 1 and Table 2 are the TCAD simulation parameters.

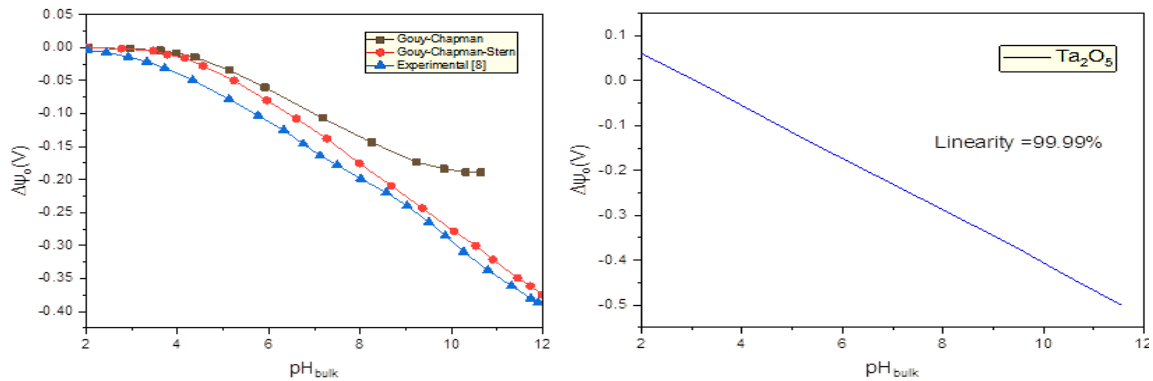


Figure 4. Theoretical surface potential shift (left) SiO<sub>2</sub> (right) Ta<sub>2</sub>O<sub>5</sub>

Table 1. TCAD Parameters of ISFET

Parameter	Value	Unit	Parameter	Value	Unit
$t_{\text{Stern}}$		-	Channel length	200	nm
T	300	K	S/D doping	$10^{17}$	$\text{cm}^{-3}$
k	$1.380649 \times 10^{-23}$	J/K	S/D length	50	nm
$t_{\text{electrolyte}}$	1000	Nm	Electrolyte concentration	$10^{-3}$	Mol/L
$t_{\text{ox}}$	3	Nm	Oxide permittivity	3.9	-
Electrolyte permittivity	80	-	$V_{\text{DS}}$	50	mV

Table 2. Materials Parameters used in TCAD Simulation

Material	Dilectric Constant	Density	$K_a$	$K_b$
SiO <sub>2</sub>	3.9	$5 \cdot 10^{14} (1/\text{cm}^2)$	$10^{-6}$	$10^2$
Ta <sub>2</sub> O <sub>5</sub>	22	$10 \cdot 10^{14} (1/\text{cm}^2)$	$10^{-4}$	$10^{-2}$

Figure 5 introduce the shifting in threshold voltage ( $V_{\text{TH}}$ ) in range of pH bulk from 2→12. indeed Shifting in threshold voltage sensitivity is the negetave values of shifting in surface potential based on the following expersion [24]:

$$\Delta V_{\text{TH}} = -\Delta \psi$$

However, the ISFET senssitivity and stability of conventional isfet for SiO<sub>2</sub> and Ta<sub>2</sub>O<sub>5</sub> as a sensing membrane contribute very well comparing with ISFET whitout Stern as shown in Figure 5. As shown the linirity of Stern-ISFET is higher than ISFET whitout Stern as well as sensivity that consistent with behaviour of Figure 4. For more analysis, the average sensvity of conventinal Stern-ISFET for Ta2O5 sensing membrane is compared with the theoratical snestivity based on Nearnst equation observed in Figure 6. As shown the Ta<sub>2</sub>O<sub>5</sub> is hit the Nearnst limit and the stability also contributed in acceptable way. The importantant aim is the linked between mathmatical model or physical interpretations and ISFET transfer charactrstics is introduced in Figure 7. In this Figure the real charactrization of pH bulk change with surface potential shifting were demostrated. As shown, the drain current is slitley change with pH range from 1→14. Consequence, the main objective of this work is achived by linking the models whitth real charactrization is clearly demostrated.

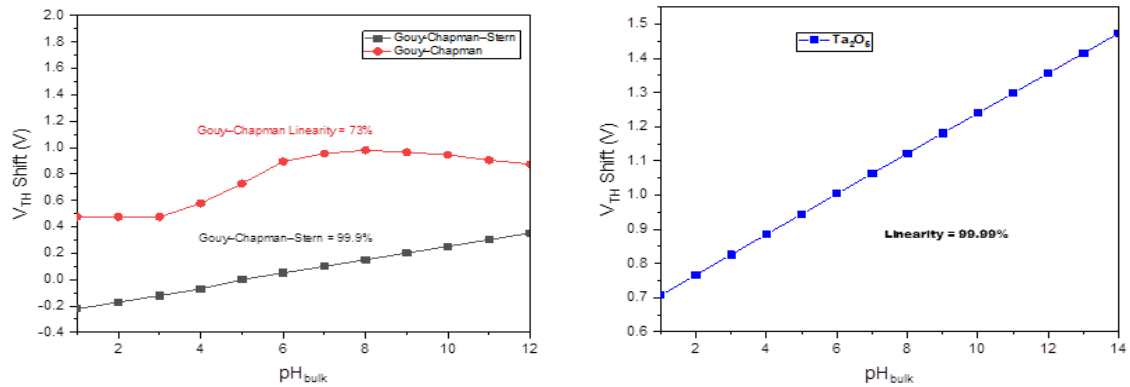


Figure 5. TCAD simulation threshold voltage shift (left) SiO<sub>2</sub> (right) Ta<sub>2</sub>O<sub>5</sub>

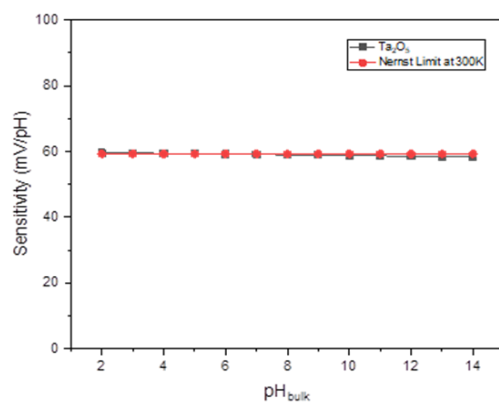


Figure 6. Average sensitivity of Ta<sub>2</sub>O<sub>5</sub> sensing membrane

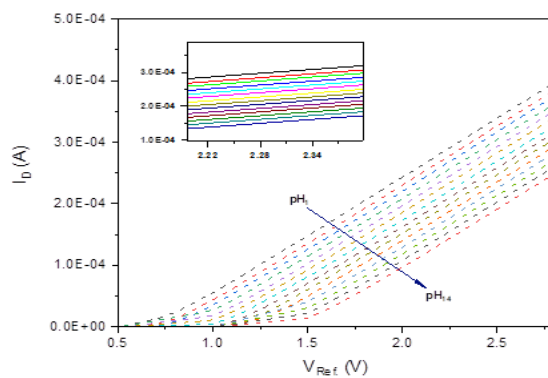


Figure 7. Conventional ISFET drain current vs. reference voltage of Ta<sub>2</sub>O<sub>5</sub> sensing membrane

For more validation, Figure 8 introduces comparative study for average sensitivity between mathematical model, TCAD simulation, Nernst Limit as a reference and other real experiments. The comparisons show excellent agreement between Model and TCAD simulation for two materials in Stern-ISFET. Despite of not good agreement with experimental value for SiO<sub>2</sub> because of lowermost of silicon dioxide, but our work achieves excellent agreement with real experimental result in Ta<sub>2</sub>O<sub>5</sub> sensing membrane as shown in Figure 8.

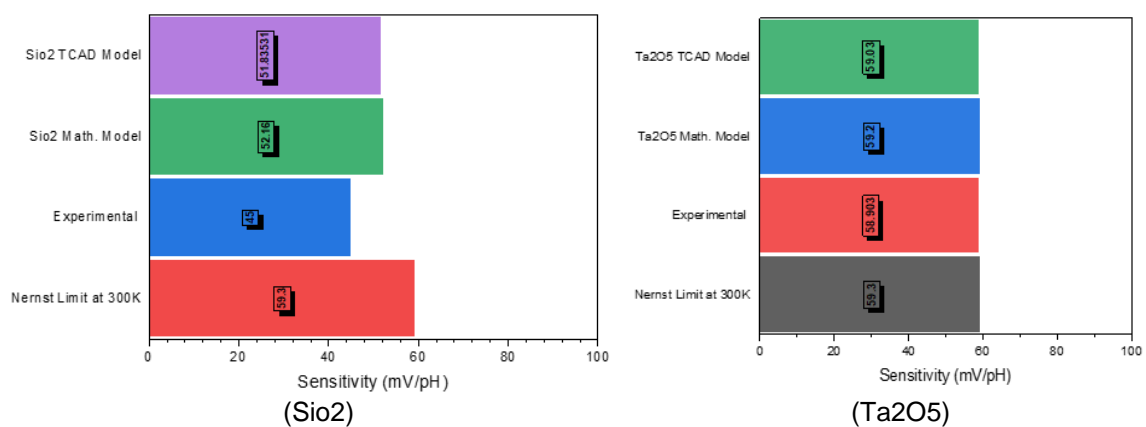


Figure 8. Conventional ISFET comparison between model, simulation, nernst limit and experimental

#### 4. Discussion

In contrast to the wide existence and importance of mathematical models for developing conventional ISFET sensitivity and ISFET concept as general, the link between these models and electrical characteristics simulation (i.e. TCAD) rather than expensive fabrication remain challenge. In this study, we adapted previous mathematical models and re-applied these models using commercial TCAD. We emphasise these models results and achieve good agreement with models as well as real experimental results for more validation. The above results show that utilizing of Gouy-Chapman-Stern model will increase the conventional ISFET sensitivity and stability of normal silicon dioxide as well as high-k materials comparing with Gouy-Chapman model. For ISFET sensitivity with  $\text{SiO}_2$  sensing membrane is raised from  $\sim 38$  mV/pH to  $\sim 51$  mV/pH, And,  $\text{Ta}_2\text{O}_5$  is hit the Nernst Limit (59.3 mV/pH) by value of 59.03 mV/pH. Furthermore, using of Stern Layer is compensate the Roll-off problem in normal model by enhancement of threshold voltage shifts linearity to reach 99.9% and 99.999% in  $\text{SiO}_2$  and  $\text{Ta}_2\text{O}_5$  sensing membranes, respectively.

Further studies should investigate the performance analysis of commonly used high-k materials using same approach. Although the simulation approach still considered not real fabrication and measurements, this study opens new directions for further analysis and optimization prior the real and cost-effective fabrication way.

#### 5. Conclusion

In this work we examine more closely the links between IC design simulation and previous mathematical models have not be clearly linked and for more accurate analysis. we adapted previous mathematical models and re-applied these models using commercial TCAD. ISFET sensitivity with  $\text{SiO}_2$  sensing membrane is raised from  $\sim 38$  mV/pH to  $\sim 51$  mV/pH, And,  $\text{Ta}_2\text{O}_5$  is hit the Nernst Limit (59.3 mV/pH) by value of 59.03 mV/pH. Furthermore, using of Stern Layer is compensate the Roll-off problem in normal model by enhancement of threshold voltage shifts linearity to reach 99.9% and 99.999% in  $\text{SiO}_2$  and  $\text{Ta}_2\text{O}_5$  sensing membranes, respectively. We anticipate this study opens new directions for further analysis and optimization prior the real and cost-effective fabrication way.

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